WHAT IS CLAIMED IS:

1. Algae-retardant roofing granules having an enhanced ability, over an extended period of time, to release algicidal metallic or bimetallic ions to prevent or inhibit the growth of algae and fungi upon atmospheric exposure of roofing surfaces coated with such granules comprising:

a) a base material of crushed mineral or synthetic aggregates in the form of granules coated with a first or inner layer and a second or outer layer;

b) said first or inner layer comprising a semi-ceramic composition consisting of a fired silicate-clay matrix containing: of from about 80 pounds to about 180 pounds of cuprous oxide per ton of said base material; or from about 5 pounds to about 40 pounds of zinc sulfide admixed with of from about 80 pounds to about 180 pounds of cuprous oxide per ton of said base material; and

c) said second or outer layer comprising a semi-ceramic composition consisting of a fired silicate-clay matrix containing: of from about 0.25 to about 2.5 pounds of an internal gas forming compound per ton of said base materials selected from the group consisting of hydrogen peroxide, alkali metal perborates, alkali metal persulfates, alkali metal borohydrides, and alkali metal azides, wherein said internal gas forming compounds form microvoids in said second or outer layer thereby rendering the second or outer layer porous to allow passage of the algicidal ions from said first or inner layer therethrough; and of from about 1 to about 20 pounds of a pigment per ton of said base material.

2. The algae-retardant roofing granules of claim 1 wherein said microvoids in said second or outer layer have an average thickness of from about 0.05 micron to about 0.5 micron.

3. The algae-retardant roofing granules of claim 1 wherein said first or inner layer contains of from about 100 to about 150 pounds of cuprous oxide per ton of base material, and from about 12 to about 25 pounds of zinc sulfide per ton of base material.

4. The algae-retardant roofing granules of claim 1 wherein said pigment is selected from the group consisting of carbon back, titanium dioxide, chromium oxide, yellow iron oxide, ultramarine blue, red iron oxide, black iron oxide, chrome titanate, and metal ferrites.

5. The algae-retardant roofing granules of claim 1 wherein said second or outer layer contains sodium perborate tetrahydrate in concert with boric acid, or a mixture of hydrogen peroxide and borax.

6. The algae-retardant roofing granules of claim 1 wherein said second or outer layer contains sodium azide or sodium borohydride.

7. The algae-retardant roofing granules of claim 1 wherein said second or outer layer optionally further comprises: of from about 80 pounds up to as high as 180 pounds of cuprous oxide per ton of said base material; or of from about 5 pounds to about 40 pounds of zinc sulfide admixed with of from about 80 pounds to about 180 pounds of cuprous oxide per ton of said base material.

8. A method of preparing algae-retardant roofing granules having an enhanced ability, over an extended period of time, to release algicidal metallic or bimetallic ions to prevent or inhibit the growth of algae or fungi upon atmospheric exposure of roofing surfaces coated with such granules comprising the steps of:

a) crushing and sizing a base mineral aggregate to form granules therefrom;

b) preheating the granules to about 210°F - 230°F;

c) coating the preheated granules with a first coat of semi-ceramic composition of an aqueous slurry comprising:
   - about 75 pounds of 38% w/w solids sodium silicate solution per ton of said base mineral aggregate,
   - about 35 pounds Kaolin clay per ton of said base mineral aggregate,
   - about 80–150 pounds cuprous oxide per ton of said base mineral aggregate,
   - about 0-25 pounds zinc sulfide per ton of said base mineral aggregate; and
   - about 0-10 pounds of pigments per ton of said base mineral aggregate;

d) pre-drying the first coated granules to a moisture content of about 0.2% to 0.5% w/w;

e) kiln-firing the pre-dried granules at a temperature of from about 740°F to about 760°F to form an insolubilized silicate-clay mixture coating in which the cuprous oxide, zinc sulfide and pigments are uniformly distributed;

f) cooling the kiln-fired, first coated granules to a temperature of from about 210°F to 230°F in preparation for application for a second or outer coating;

g) coating the cooled granules with a second coat of a semi-ceramic composition of an aqueous slurry comprising:
   - about 40 pounds sodium silicate per ton of said base mineral aggregate,
   - about 25 pounds of Kaolin clay per ton of said base mineral aggregate,
   - about 0.5 – 1.5 of an internal gas forming compound selected from the group consisting of hydrogen peroxide, alkali metal perborates, alkali metal persulfates, alkali metal borohydrides, and alkali metal azides per ton of said base mineral aggregates, said internal gas forming compounds forming microvoids thereby rendering the coating porous to allow passage of said algicidal ions from said inner coat therethrough,

- about 0.5 – 1.5 pounds of a solubilizer/stabilizer per ton of said base mineral aggregate, and
- about 0 – 15 pounds of a pigment per ton of said base mineral aggregate;

h) pre-drying the second coated granules to a moisture content of about 0.2 to 0.5% w/w;

i) kiln-firing the granules at a temperature of from about 890°F to about 910°F to form an insolubilized silicate-clay matrix;

j) reducing the temperature of the granules to about 200° to 220°F; and

k) treating the granules with a mixture of processed oil and an organosilicone compound to impart dust control and adhesion to a substrate.

9. A method of protecting asphalt-coated roof shingles against algae and fungi comprising the steps of:

a) providing asphalt coated roof shingles;

b) providing a base mineral aggregate and forming granules therefrom;

c) coating said granules with a first layer of a semi-ceramic composition consisting of a fired silicate-clay matrix containing: of from about 80-pounds to about 180 pounds of cuprous oxide per ton of said base material; or from about 5 pounds to about 40 pounds of zinc sulfide admixed with of from about 80 pounds to about 180 pounds of cuprous oxide per ton of said base material;

d) coating said first layer coated granules with a second layer of a semi-ceramic composition consisting of a fired silicate-clay matrix containing:

- of from about 0.5 to about 1.5 pounds of an internal gas-forming compound per ton of said base materials selected from the group consisting of hydrogen peroxide, alkali metal perborates, alkali metal persulfates, alkali metal borohydrides, and alkali metal azides wherein said internal gas-forming compounds form microvoids in said second

or outer layer thereby rendering the second or outer layer porous to allow passage of said algaecidal ions from said first layer therethrough,

- about 56 pounds of 40% w/w solids sodium silicate solution per ton of said base mineral aggregate,
- about 25 pounds of Kaolin clay per ton of said base mineral aggregate,
- about 0.5 – 1.5 pounds of a solubilizer/stabilizer per ton of said base mineral aggregate, and
- about 0-15 pounds of a pigment per ton of said base mineral aggregate;

e) treating said first and second layer coated granules with a mixture of process oil and an organosilicone compound to impart dust control and adhesive properties thereto to obtain treated granules ready for coating said asphalt coated roof shingles; and

f) applying said treated granules to said roofing shingles to obtain algae and fungi-resistant roof shingles.